Random gap model for graphene and graphene bilayers

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The effect of a randomly fluctuating gap, created by a random staggered potential, is studied in a monolayer and a bilayer of graphene. The density of states, the one-particle scattering rate and transport properties (diffusion coefficient and conductivity) are calculated at the neutrality point. All these quantities vanish at a critical value of the average staggered potential, signaling a continuous transition to an insulating behavior. The calculations are based on the self-consistent Born approximation for the one-particle scattering rate and a massless mode of the two-particle Green's function which is created by spontaneous symmetry breaking. Transport quantities are directly linked to the one-particle scattering rate. Moreover, the effect of disorder is very weak in the case of a monolayer but much stronger in bilayer graphene.

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Graphene, a sheet of carbon atoms, or bilayer graphene are semimetals with good conducting properties [1, 2, 3]. In particular, the minimal conductivity at the neutrality point (NP) is very robust and almost unaffected by disorder or thermal fluctuations [3, 4, 5, 6]. Recent experiments with hydrogenated graphene [7] and biased bilayer graphene [8, 9, 10] have revealed that a staggered potential (SP) can be created in graphene and bilayer graphene which breaks the sublattice symmetry. This opens a gap at the Fermi energy, leading to an insulating behavior. With this opportunity one enters a new field, where one can switch between a conducting and an insulating regime of a two-dimensional material, either by a chemical process (e.g. oxidation or hydrogenation) or by applying an external electric field [11].

It is clear that the opening of a uniform gap destroys the metallic state immediately. This means that the (minimal) conductivity at the NP drops from a finite value of order e^2/h directly to zero. In a realistic system, however, the gap may not be uniform. This means that locally gaps open, whereas in other regions of the sample there is no gap. The situation can be compared with a classical random network of broken and unbroken bonds. The conductivity of such a network is nonzero as long as there is a percolating cluster of unbroken bonds. In such a system the transition from conducting to insulting behavior is presumably a second order percolation transition [12].

Disorder in graphene has been the subject of a number of recent numerical studies [13, 14]. The results can be summarized by the statement that chiral-symmetry preserving disorder provides delocalized states whereas a chiral-symmetry breaking potential disorder leads to Anderson localization, even at the NP.

Conductivity and other transport properties in graphene can be evaluated by solving the Bethe-Salpeter equation for the average two-particle Green's function (Cooperon) [15, 16, 17, 18, 19]. Unfortunately, the Bethe-Salpeter equation is usually a complex matrix equation which is difficult to handle. Therefore, a different approach will be employed here that eliminates a part of the complexity by focusing on continuous symmetries and spontaneous symmetry breaking. This allows us to identify a (massless) diffusion mode in the system with a randomly fluctuating gap. Consequently, diffusion can only stop when the spontaneous symmetry breaking vanishes. It will be discussed in this paper that this can happen if the average SP approaches a critical value. Moreover, there is no drop of the conductivity but a continuous decay to zero, depending on the fluctuations of the SP.

model: Quasiparticles in monolayer graphene (MLG) or bilayer graphene (BLG) are described in tight-binding approximation by a nearest-neighbor hopping Hamiltonian

$$\mathbf{H} = -t \sum_{<r,r'>} c_r^\dagger c_{r'} + \sum_r m_r c_r^\dagger c_r + h.c. \ , \qquad (1)$$

where the underlying structure is either a honeycomb lattice (MLG) or two honeycomb lattices with Bernal stacking (BLG).

The sublattice symmetry of the honeycomb lattice is broken by a staggered potential (SP) m_r which is positive (negative) on sublattice A (B) [20, 21]. Such a potential can be the result of chemical absorption of other atoms (e.g. oxygen or hydrogen [7]) or of an external gate voltage applied to the two layers of BLG [8]. Neither in MLG nor in BLG the potential m_r and, therefore, the gap is uniform, because of fluctuations in the coverage of the MLG by additional non-carbon atoms or by the fact that the graphene sheets are not planar [22, 23, 24]. Deviations from the planar structure in the form of ripples cause fluctuations in the distance of the two sheets in BLG which results in an inhomogeneous potential m_r along each sheet. It is assumed that the gate voltage is adjusted at the NP such that in average the SP is exactly antisymmetric: $\langle m_A \rangle = -\langle m_B \rangle$.

At first glance, the Hamiltonian in Eq. (1) is a standard hopping Hamiltonian with random potential m, fre-

quently used to study the generic case of Anderson localization [25]. The dispersion, however, is special in the case of graphene due to the honeycomb lattice: at low energies it consists of two valleys K and K' [18, 23]. It is assumed that weak disorder scatters only at small momentum such that intervalley scattering is not relevant. Then each valley contributes separately to transport, and the contribution of the two valleys to the conductivity σ is additive: $\sigma = \sigma_K + \sigma_{K'}$. This allows us to consider for the low-energy properties a Dirac-type Hamiltonian for each valley separately

$$H = h_1\sigma_1 + h_2\sigma_2 + m\sigma_3 \tag{2}$$

with Pauli matrices σ_j and with h_j

$$h_j = i\nabla_j \ \ (MLG), \ \ h_1 = \nabla_1^2 - \nabla_2^2, \ h_2 = 2\nabla_1\nabla_2 \ \ (BLG) \ . \tag{3}$$

∇_j is the lattice difference operator in j $(= 1, 2)$ direction. Within this approximation the SP m_r is a random variable with mean value $\langle m_r\rangle_m = \bar{m}$ and variance $\langle (m_r - \bar{m})(m_{r'} - \bar{m})\rangle_m = g\delta_{r,r'}$. The following transport calculations will be based entirely on the Hamiltonian of Eq. (2). In particular, the average Hamiltonian $\langle H\rangle_m$ can be diagonalized by Fourier transformation and is $k_1\sigma_1 + k_2\sigma_2 + \bar{m}\sigma_3$ for MLG with eigenvalues $E_k = \pm\sqrt{\bar{m}^2 + k^2}$. For BGL the average Hamiltonian is $(k_1^2 - k_2^2)\sigma_1 + 2k_1k_2\sigma_2 + \bar{m}\sigma_3$ with eigenvalues $E_k = \pm\sqrt{\bar{m}^2 + k^4}$.

symmetries: Transport properties are determined by the model properties on large scales. The latter are controlled by the symmetry of the Hamiltonian and of the corresponding one-particle Green's function $G(i\epsilon) = (H + i\epsilon)^{-1}$. In the absence of sublattice-symmetry breaking (i.e. for $m = 0$), the Hamiltonian $H = h_1\sigma_1 + h_2\sigma_2$ has a continuous chiral symmetry

$$H \to e^{\alpha\sigma_3} H e^{\alpha\sigma_3} = H \tag{4}$$

with a continuous parameter α, since H anticommutes with σ_3. The SP term $m\sigma_3$ breaks the continuous chiral symmetry. However, the behavior under transposition $h_j^T = -h_j$ for MLG and $h_j^T = h_j$ for BLG provides a discrete symmetry:

$$H \to -\sigma_j H^T \sigma_j = H \ , \tag{5}$$

where $j = 1$ for MLG and $j = 2$ for BLG. This symmetry is broken for the one-particle Green's function $G(i\epsilon)$ by the $i\epsilon$ term. To see whether or not the symmetry is recovered for $\epsilon \to 0$, the difference

$$G(i\epsilon) + \sigma_j G^T(i\epsilon)\sigma_j = G(i\epsilon) - G(-i\epsilon) = i\pi\rho(E = 0) \tag{6}$$

must be evaluated, where $\rho(E = 0) \equiv \rho_0$ is the density of states at the NP. Here the limit $\epsilon \to 0$ is implicitly assumed. Thus the order parameter for spontaneous symmetry breaking is ρ_0.

conductivity: The conductivity can be calculated from the Kubo formula. Here we focus on interband scattering between states of energy $\omega/2$ and $-\omega/2$, which is a major contribution to transport near the NP. The frequency-dependent conductivity then reads [26]

$$\sigma_0(\omega) = -\frac{e^2}{2h}\omega^2 \langle\langle \Phi_{-\omega/2}|r_k^2|\Phi_{\omega/2}\rangle\rangle_m \ , \tag{7}$$

where $|\Phi_E\rangle$ is an eigenstate of H in Eq. (2) with energy E. In other words, the conductivity is proportional to a matrix element of the position operator r_k $(k = 1, 2)$ with respect to energy eigenfunctions from the lower and the upper band. The matrix element $\langle\Phi_{\omega/2}|r_k^2|\Phi_{-\omega/2}\rangle$ is identical with the two-particle Green's function

$$\sum_r r_k^2 Tr_2 \left[G_{r0}(-\omega/2 - i\epsilon)G_{0r}(\omega/2 + i\epsilon)\right] \ . \tag{8}$$

This indicates that transport properties are expressed by the two-particle Green's function $G(i\epsilon)G(-i\epsilon)$. Each of the two Green's functions, $G(i\epsilon)$ and $G(-i\epsilon)$, can be considered as a random variable which are correlated due to the common random variable m_r. Their distribution is defined by a joint distribution function $P[G(i\epsilon), G(-i\epsilon)]$. In terms of transport theory, both Green's functions must be included on equal footing. This is possible by introducing the extended Green's function

$$\hat{G}(i\epsilon) = \begin{pmatrix} G(i\epsilon) & 0 \\ 0 & G(-i\epsilon) \end{pmatrix} = \begin{pmatrix} H + i\epsilon & 0 \\ 0 & H - i\epsilon \end{pmatrix}^{-1} \ .$$

In the present case one can use the symmetry transformation of H in Eq. (5) to write the extended Green's function as

$$\begin{pmatrix} \sigma_0 & 0 \\ 0 & i\sigma_j \end{pmatrix} \begin{pmatrix} H + i\epsilon & 0 \\ 0 & H^T + i\epsilon \end{pmatrix}^{-1} \begin{pmatrix} \sigma_0 & 0 \\ 0 & i\sigma_j \end{pmatrix} \ .$$

This introduces an extended Hamiltonian $\hat{H} = diag(H, H^T)$ which is invariant under a global "rotation"

$$\hat{H} \to e^S \hat{H} e^S = \hat{H} \ , \quad S = \begin{pmatrix} 0 & \alpha\sigma_j \\ \alpha'\sigma_j & 0 \end{pmatrix} \tag{9}$$

with continuous parameters α, α', since $\hat{H}$ anticommutes with S. The $i\epsilon$ term of the Green's function also breaks this symmetry. According to Eq. (6), the symmetry is broken spontaneously for $\epsilon \to 0$ if the density of states ρ_0 is nonzero. Since this is a continuous symmetry, there is a massless mode which describes diffusion [27]. Symmetry breaking should be studied for average quantities. Therefore, the average density of states must be evaluated.

spontaneous symmetry breaking: The average one-particle Green's function can be calculated from the average Hamiltonian $\langle H\rangle_m$ by employing the self-consistent Born approximation (SCBA) [15, 16, 21]

$$\langle G(i\epsilon)\rangle_m \approx (\langle H\rangle_m - 2\Sigma)^{-1} \equiv G_0(i\eta, m_s) \ . \tag{10}$$

The self-energy Σ is a 2×2 tensor due to the spinor structure of the quasiparticles: $\Sigma = -(i\eta\sigma_0 + m_s\sigma_3)/2$. Scattering by the random SP produces an imaginary part of the self-energy η (i.e. a one-particle scattering rate) and a shift m_s of the average SP $\bar{m}$ (i.e., $\bar{m} \to m' \equiv \bar{m} + m_s$). Σ is determined by the self-consistent equation

$$\Sigma = -g\sigma_3(\langle H \rangle_m - 2\Sigma)^{-1}_{rr}\sigma_3 \ . \tag{11}$$

For simplicity, the dc limit $\omega \sim 0$ is considered here. The average density of states at the NP is proportional to the scattering rate: $\rho_0 = \eta/2g\pi$. This reflects that scattering by the random SP creates a nonzero density of states at the NP. It should be noticed that the entire calculation of the one-particle scattering rate η is based on the average one-particle Green's function. Therefore, it is unrelated to the continuous symmetry of Eq. (9). On the other hand, $\eta > 0$ implies spontaneous breaking of this symmetry.

Eq. (11) can also be written in terms of two equations, one for the one-particle scattering rate η and another for the shift of the SP m_s, as

$$\eta = gI\eta, \quad m_s = -\bar{m}gI/(1+gI) \ . \tag{12}$$

I is a function of $\bar{m}$ and η and also depends on the Hamiltonian. For MLG it reads with momentum cutoff λ

$$I = \frac{1}{2\pi} \ln\left[1 + \frac{\lambda^2}{\eta^2 + (\bar{m}+m_s)^2}\right] \tag{13}$$

and for BLG

$$I \sim \frac{1}{4\sqrt{\eta^2 + (\bar{m}+m_s)^2}} \quad (\lambda \sim \infty) \ . \tag{14}$$

A nonzero solution η requires $gI = 1$ in the first part of Eq. (12), such that $m_s = -\bar{m}/2$ from the second part. Since the integrals I are monotonically decreasing functions for large $\bar{m}$, a real solution with $gI = 1$ exists only for $|\bar{m}| \leq m_c$. For both, MLG and BLG, the solutions read

$$\eta^2 = (m_c^2 - \bar{m}^2)\Theta(m_c^2 - \bar{m}^2)/4 \ , \tag{15}$$

where the model dependence enters only through the critical average SP m_c:

$$\frac{2\lambda}{\sqrt{e^{2\pi/g}-1}} \sim 2\lambda e^{-g/\pi} \ \ (MLG), \quad g/2 \ \ (BLG) \ . \tag{16}$$

m_c is much bigger for BGL (cf. Fig. 1), a result which indicates that the effect of disorder is much stronger in BLG. This is also reflected by the scattering rate at $\bar{m} = 0$ which is $\eta = m_c/2$.

diffusion: The average two-particle Green's function

$$K^{-1}_{rr'}(i\epsilon) = -\langle Tr_2[G_{rr'}(-i\epsilon)G_{r'r}(i\epsilon)]\rangle_m$$

can be evaluated from an effective field theory [27]. If $\eta > 0$ the corresponding spontaneous breaking of the symmetry in Eq. (9) creates one massless mode, which is related to a diffusion propagator in Fourier space:

$$\frac{1}{K_q(i\epsilon)} \sim -\frac{\eta/g}{\epsilon + Dq^2}$$

with the diffusion coefficient

$$D = g\frac{\eta}{2}\sum_r r_k^2 Tr_2[G_{0,r0}(-i\eta)G_{0,0r}(i\eta)] \ . \tag{17}$$

Within this approximation the matrix element of the position operator reads

$$\langle\langle \Phi_{\omega/2}|r_k^2|\Phi_{-\omega/2}\rangle\rangle_m = \frac{\partial^2}{\partial q_k^2}\frac{1}{K_q(\omega/2)}\Big|_{q=0} \sim -8\frac{\eta}{g\omega^2}D \ . \tag{18}$$

Using the relation between the matrix element and the two-particle Green's function in Eq. (8), the diffusion coefficient becomes $D = (g\eta/2)\langle\Phi_{i\eta}|r_k^2|\Phi_{-i\eta}\rangle$. Inserting this on the right-hand side of Eq. (18) gives a simple relation between the disorder averaged matrix element of r_k^2 and the corresponding matrix element without disorder:

$$\langle\langle \Phi_{\omega/2}|r_k^2|\Phi_{-\omega/2}\rangle\rangle_m = -\frac{\eta^2}{(\omega/2)^2}\langle\Phi_{i\eta}|r_k^2|\Phi_{-i\eta}\rangle \ . \tag{19}$$

This is similar to the relation of the average one-particle Green's function in the SCBA of Eq. (10). Like in the latter case, the averaging process leads to a change of energies $\omega/2 \to i\eta$ (i.e. a replacement of the frequency by the scattering rate). Moreover, in the relation of the two-particle Green's function there is an extra prefactor $-\eta^2/(\omega/2)^2$. It is important for the transport properties, since the average matrix element diverges like ω^{-2}. This indicates that the states $|\Phi_{\pm\omega/2}\rangle$ are delocalized for $\omega = 0$ in the presence of weak SP disorder, and localization increases as one goes away from the NP. Such a behavior was also found for bond disorder in analytic [26] and in numerical studies [14].

After evaluating $\langle\Phi_{i\eta}|r_k^2|\Phi_{-i\eta}\rangle$, the results for the diffusion coefficient in Eq. (17) and for the conductivity in Eqs. (7), (18) can be summerized in the following expressions

$$D \sim \frac{ag}{4\pi}\frac{\eta}{\eta^2 + \bar{m}^2/4} \ , \quad \sigma_0 \sim \frac{a}{\pi}\frac{\eta^2}{\eta^2 + \bar{m}^2/4}\frac{e^2}{h} \ , \tag{20}$$

where $a = 1$ ($a = 2$) for MLG (BLG). First, this result indicates that the physical relevant quantity is the one-particle scattering rate η. The difference between MLG and BLG is only due to the parameter $a = 1, 2$ and due to the $\bar{m}$-dependent scattering rate η. Second, the result reflects a diffusive behavior as long as the scattering rate η does not vanish. Eq. (15) gives a vanishing scattering rate for $\bar{m} = m_c$, where the critical value m_c is twice the scattering rate at $\bar{m} = 0$. Moreover, the average density of states at the NP is proportional to η. Therefore, a

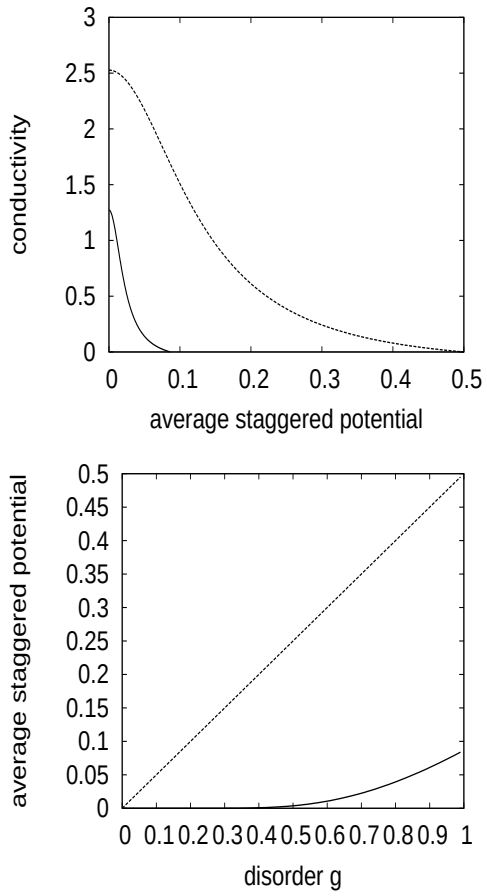

FIG. 1: upper panel: dc conductivity in units of e^2/h for BL graphene (upper curve) and ML graphene (lower curve) vs. the average staggered potential $\bar{m}$, calculated from Eq. (20) for $g = 1$ and $\lambda = 1$. lower panel: critical average staggered potential as a function of g (variance of the staggered potential fluctuations) for BL graphene (upper curve) and ML graphene (lower curve).

global gap opens only for $\bar{m} > m_c$. Details of the transport properties distinguish between $\bar{m} = 0$ and $\bar{m} \neq 0$.

$\bar{m} = 0$: A fluctuating SP with g not too large has no effect on the conductivity. This can also be understood from the Einstein relation $\sigma_0 \propto D\rho$, since the density of states ρ is proportional and the diffusion coefficient D is inversely proportional to the normalized scattering rate η/g. Such a behavior was also observed in the chiral-invariant case with random bond disorder which is related to ripples [3, 16, 22, 26]. The scattering rate η increases with disorder strength g (cf. Fig. 1). Consequently, the density of states at the NP $\rho_0 = \eta/2g\pi$ increases with g for MLG, at least for small values of g, whereas it is constant for BLG. On the other hand, the diffusion coefficient decreases with g/η, as a result of increased scattering.

$\bar{m} \neq 0$: The conductivity decreases with $\bar{m}$ and eventually goes to zero at $\bar{m} = m_c$. This is due to two effects, namely the reduction of the density of states and the reduction of the diffusion coefficient with $\bar{m}$, caused by a fluctuating gap. Since the product of the two quantities give the conductivity in the Einstein relation, the conductivity also decreases.

The only difference between MLG and BLG in our calculation is the linear (MLG) and the quadratic (BLG) spectrum. This has quantitative consequences for the conductivity: For BLG it is twice as big as for MLG at $\bar{m} = 0$ and also decays on a larger scale for $0 < \bar{m} \leq m_c$, since the critical value is $m_c = g/2$ for BLG, whereas it is $m_c \sim \exp(-\pi/g)$ for MLG. As shown in Fig. 1, the conductivity of MLG vanishes at much lower values of $\bar{m}$. Remarkable is the enormous difference of the scattering rate between the two systems at $\bar{m} = 0$. As shown in Fig. 1, η is practically zero for a large interval of g, whereas it increases linearly with g for BLG. This indicates that disorder has a much stronger effect in the latter.

Our result of the random SP represents a case that is different from random bond disorder (i.e. with chiral symmetry) and random scalar potential (breaks the chiral symmetry but not the sublattice symmetry). The former does not localize states at the NP, whereas the latter has presumably always localized states, with a very large localization length though. In a recent paper, Zhang et al. suggested a Kosterlitz-Thouless (KT) transition for a long-range random potential [13]. The KT transition is a phase transition that has no spontaneous symmetry breaking but a single massless mode in the ordered phase due to $U(1)$ phase fluctuations. In the case of the random SP the situation is very different: There is spontaneous symmetry breaking in the diffusive phase due to $\eta > 0$. Moreover, the symmetry of the fluctuations in Eq. (9) has two components rather than one. Therefore, the transition to the insulating behavior due to random SP cannot be linked to the conventional KT transition.

A possible experimental realization of a random gap was recently observed by Adam et al. [28]. It still remains to be seen whether or not the observed transition, which was studied by varying the gate voltage at a fixed gap, can be related to a nonzero average SP. This would require a tuning of the gap fluctuations and measurement of the local density of states.

In conclusion, the one-particle scattering rate, the density of states, the diffusion coefficient, and the conductivity decrease with increasing average SP $\bar{m}$ and vanish at a critical point m_c. The latter is exponentially small for MLG but proportional to disorder strength for BLG. Thus the effect of disorder is much stronger in BLG.

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